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The dynamics of electron gratings formed in $Hg_{1-x}Cd_xTe$ at 80°K by the interference of pump and probe beams from a CO_2 TEA laser has been investigated. A CO_2 TEA laser beam incident upon the backside of the sample quenches the forward mode phase conjugate signal from the pump and probe beams, with a response time no greater than 40 nsec. A model has been proposed based upon two-photon absorption and Auger recombination. A theoretical analysis of $GaAs/Al_xGa_{1-x}As$ and $Hg_{1-x}Cd_xTe/Hg_yCd_{1-y}Te$ superlattices shows that the photo-excited plasma mechanism does not give rise to an appreciably larger third order susceptibility than bulk alloys. However, the third order susceptibility arising from conduction band nonparabolicity can be two orders of magnitude higher in the $GaAs/Al_xGa_{1-x}As$ superlattices than in the bulk alloy. There is no increase in the third order susceptibility due to conduction band nonparabolicity in $Hg_{1-x}Cd_xTe/Hg_yCd_{1-y}Te$ superlattices compared with the bulk alloy. One paper was presented at a scientific meeting and one was submitted for publication to a technical journal.

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1.0 RESEARCH OBJECTIVES

The objectives of the contract are listed below:

(1) Determine the dependence of the power reflection coefficient upon signal and pump intensities for optical phase conjugation by resonant four-wave mixing in mercury cadmium telluride crystals.

(2) Study optical phase conjugation by four-wave mixing in epitaxial layers of mercury cadmium telluride.

(3) Investigate noncollinear phase matched far infrared radiation in mercury cadmium telluride.

(4) Measure the spectral dependence of the optical absorption coefficient in mercury cadmium telluride from 10 to 50 micrometers and separate band edge absorption with possible exciton effects from intervalence band and free carrier absorption.

(5) Measure the spectral dependence of the quantum efficiency in small gap mercury cadmium telluride from 10 to 50 micrometers.

(6) Determine the relative contributions of the microscopic mechanisms, including conduction band nonparabolicity, photoexcited plasma, and saturable absorption, to optical phase conjugation in $Hg_{1-x}Cd_xTe$.

(7) Investigate the quality of the phase conjugate return in $Hg_{1-x}Cd_xTe$.

(8) Investigate optical bistability in $Hg_{1-x}Cd_xTe$ arising from third order nonlinearities.

(9) Investigate theoretically the response time of nonlinear optical interactions produced by the various microscopic mechanisms in semiconductors.

(10) Investigate theoretically the nonlinear optical interaction mechanisms in semiconductor superlattices.

2.0 STATUS OF RESEARCH EFFORT AND FUTURE PLANS

2.1 Nonlinear Optics Experimental Investigations - Status

During the period 9 July 1984 - 8 January 1985 the nonlinear optics experimental investigations continued to concentrate on the dynamics of real-time

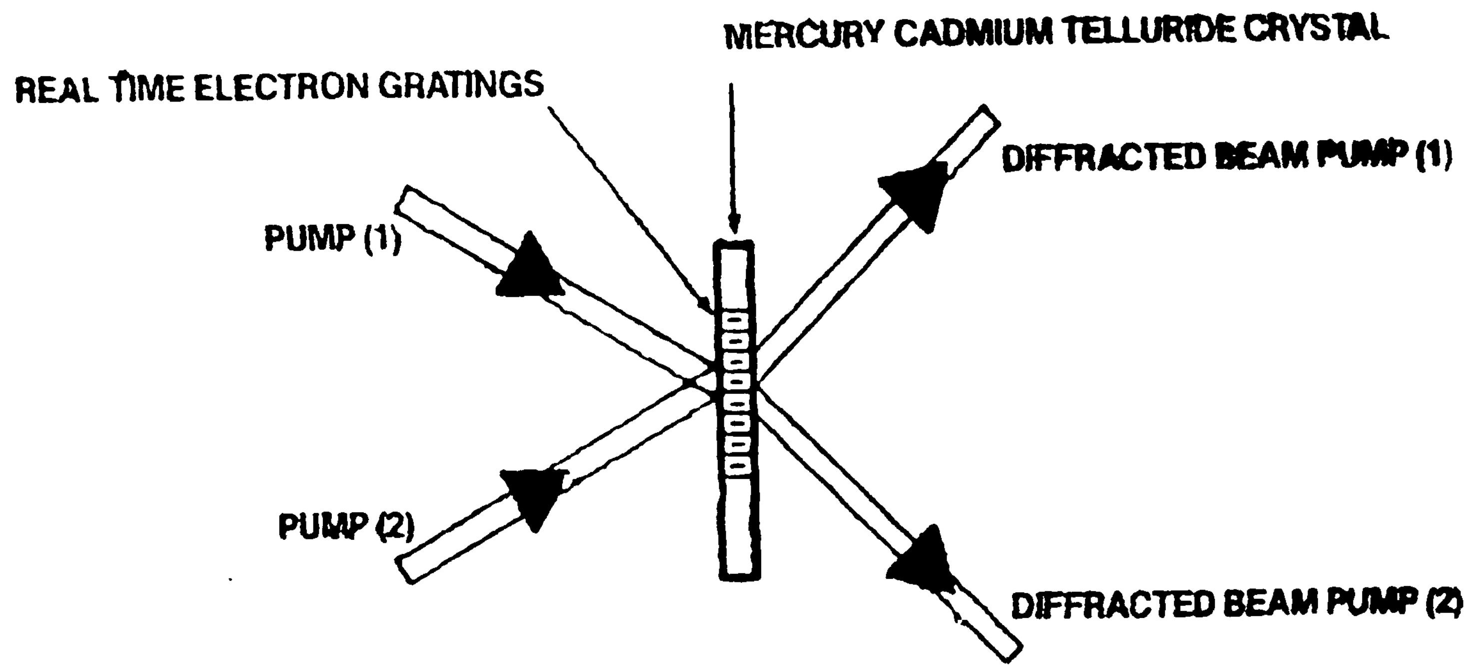
electron gratings formed in $Hg_{1-x}Cd_xTe$ by the interference of two CO_2 laser beams. These experiments, which began during the period 9 July 1983 - 8 January 1984 (see report number 6) were continued during the subsequent reporting period (see report number 7) and the present period. Some of the earlier results are summarized below, together with new data. These were the subject of a paper entitled "Dynamics of Real Time Electron Gratings in $Hg_{1-x}Cd_xTe$", by M.A. Khan, J. Lehman, and P.W. Kruse, which was presented at the International Conference on Lasers '84, San Francisco, November 26-30, 1984.

The interference of two CO_2 laser beams in a $Hg_{1-x}Cd_xTe$ crystal produces a real-time electron grating, see Figure 1. Each of the beams is diffracted by the grating as shown in the figure. An alternative, equally valid, description is that the inteference of the two incident beams forms two forward mode phase conjugate signals.

The experimental arrangement for investigating these effects is shown in Figure 2. Here the more intense incident beam is referred to as the pump; the weaker is the probe. The incident beams are obtained by beam splitting the output from either a CO_2 TEA laser or a CO_2 Q-switched laser. The forward mode phase conjugate signal arising from diffraction of the pump beam from the crystal is detected by a Ge:Cu detector, amplified, and displayed on an oscilloscope.

Experiments of this type, described in report number 6, revealed that the phase conjugate forward mode signal could be quenched by a beam incident upon the back side of the crystal. Although the beams incident upon the front side must be parallel-polarized, it was found that the effect produced by the beam incident upon the back side was polarization insensitive. The beam incident upon the back side is termed the "erase" beam. This quenching of the forward mode phase conjugate signal by a randomly polarized signal from the back is a novel effect, not reported elsewhere.

During the present reporting period the dependence of the forward mode phase conjugate signal upon erase beam intensity was determined. It was found that the erase beam need not be obtained by beam splitting the CO_2 laser which is the source of the pump and probe beam. An experiment was carried out in which a CO_2 TEA laser



- LIGHT DIFFRACTION FROM REAL TIME ELECTRON GRATINGS
- DIFFRACTION EFFICIENCY IS A MEASURE OF $\chi^{(3)}$
- EQUIVALENCE TO REAL TIME FOUR WAVE MIXING

FIGURE 1

Real-Time Electron Grating

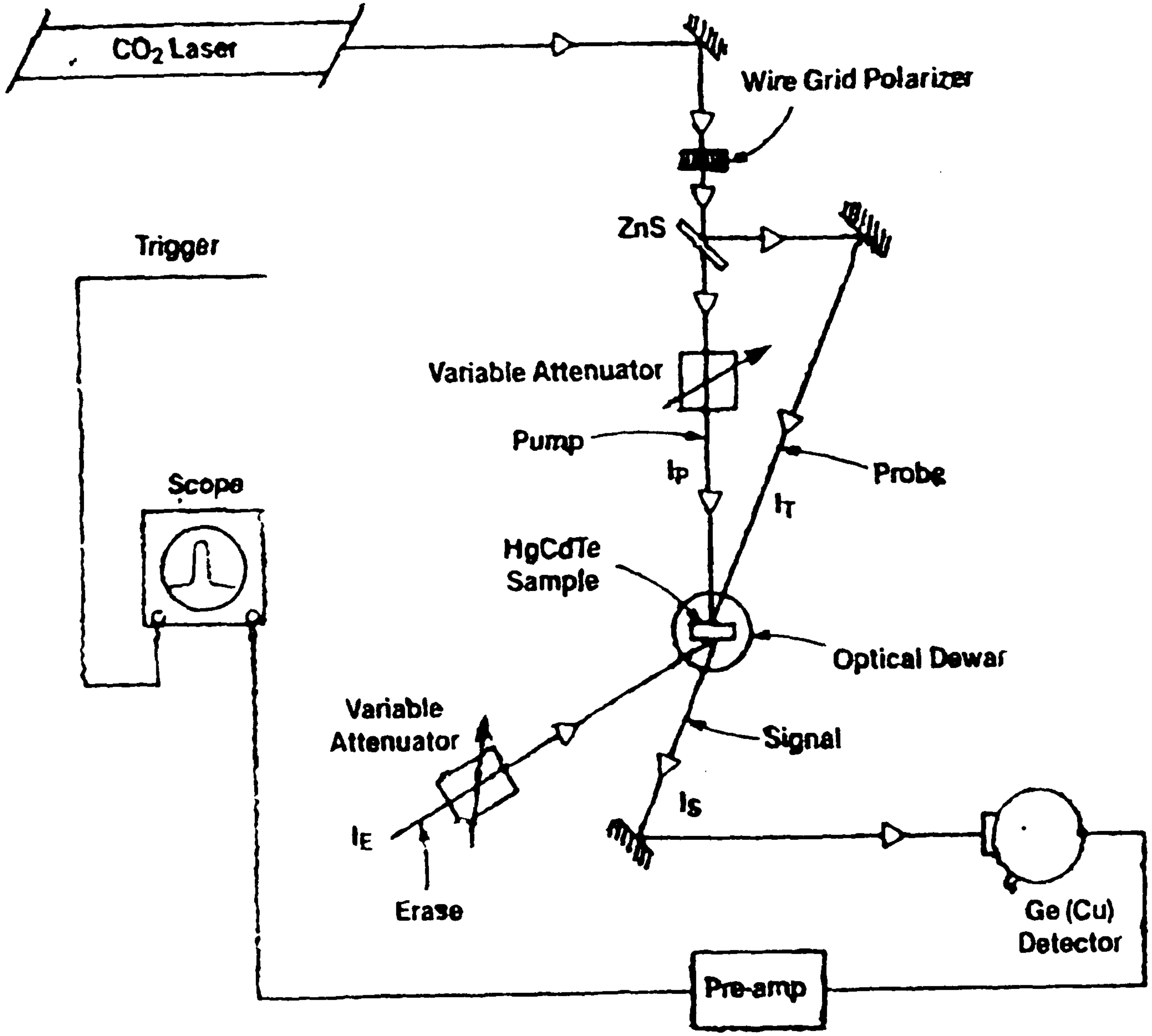


FIGURE 2

Experimental Arrangement

was the source of the pump and probe beams, and a second CO_2 TEA laser was the source of the erase beam. Results were similar to those in which one laser was used. Thus coherence between the erase beam and the pump and probe beams is not required. It was also determined that erasing was independent of the angle which the erase beam made with the back side of the sample.

The dependence of the quenching effect upon erase beam intensity was determined during this reporting period. A sample of n-type $Hg_{0.77}Cd_{0.23}Te$ was employed in an optical Dewar at 80^oK. A TEA CO_2 laser was the source of the pump, probe, and erase beams. Figure 3 shows that as the erase beam intensity was increased, the diffracted signal intensity decreased rapidly, then continued to decrease at a much slower rate.

The grating relaxation time was also studied during this period. Here a time delay was introduced between the onset of the probe and pump TEA laser pulses and the onset of the TEA laser erase pulse. This was done by causing the erase pulse, which was formed by beam splitting the TEA laser output, to traverse an adjustable path length before it was incident upon the back side of the $Hg_{0.77}CD_{0.23}Te$ sample at 80^oK. As illustrated in Figure 4, the largest erasure occured when the erase pulse was incident simultaneously with the pump and probe pulses. As the time delay was increased,the effect was reduced, until it disappeared at a delay of 40 nsec.

The data of Figures 3 and 4 were obtained when the erase beam completely overlapped the pump and probe beams. During this period it was determined that when the erase beam fills only half of the pump/probe beam areas, the diffracted signal is only reduced by a factor of two. A more detailed study using a knife edge, see Figure 5, showed that the magnitude of the erase effect is directly proportional to the overlap between the erase beam and the pump and probe beams.

During this reporting period, an attempt has been made to develop a model which explains the effect of the erase beam. The outline of the model is as follows:

- Production of hole-electron plasma by two-photon absorption.

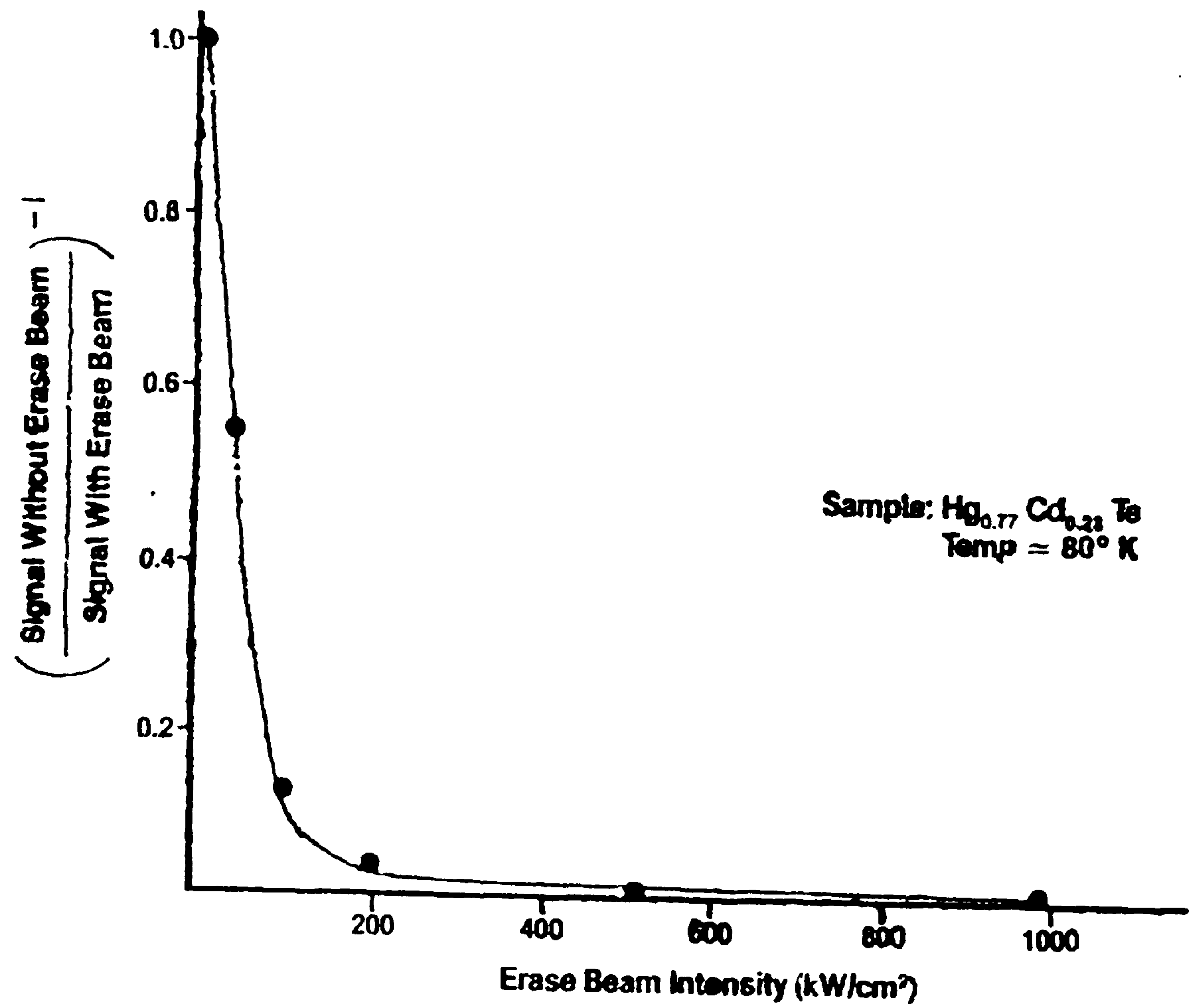


FIGURE 3

Dependence of Diffracted Beam Signal Upon Erase Beam Intensity

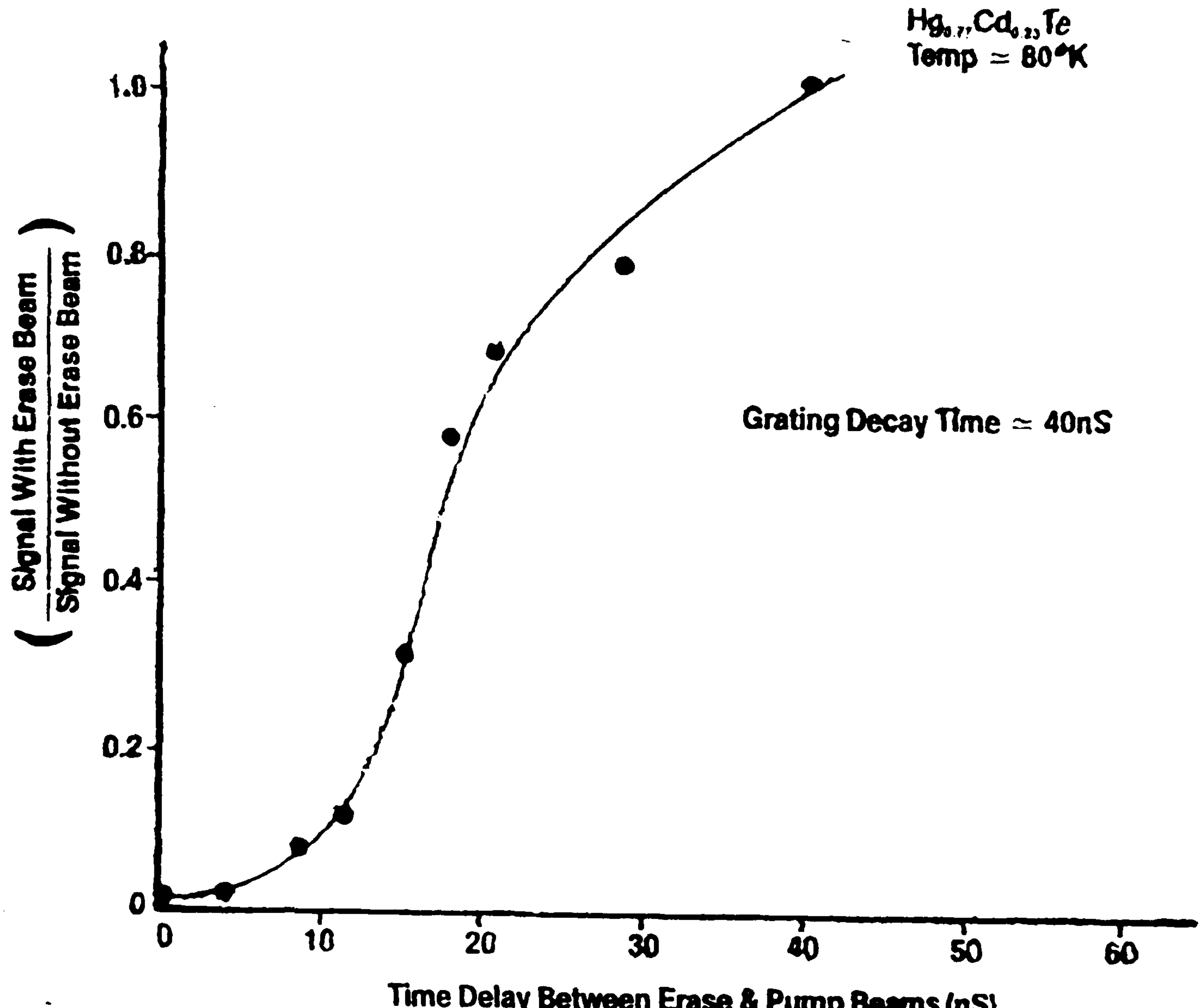


FIGURE 4

Dependence of Diffracted Beam Signal Upon Erase Beam Time Delay

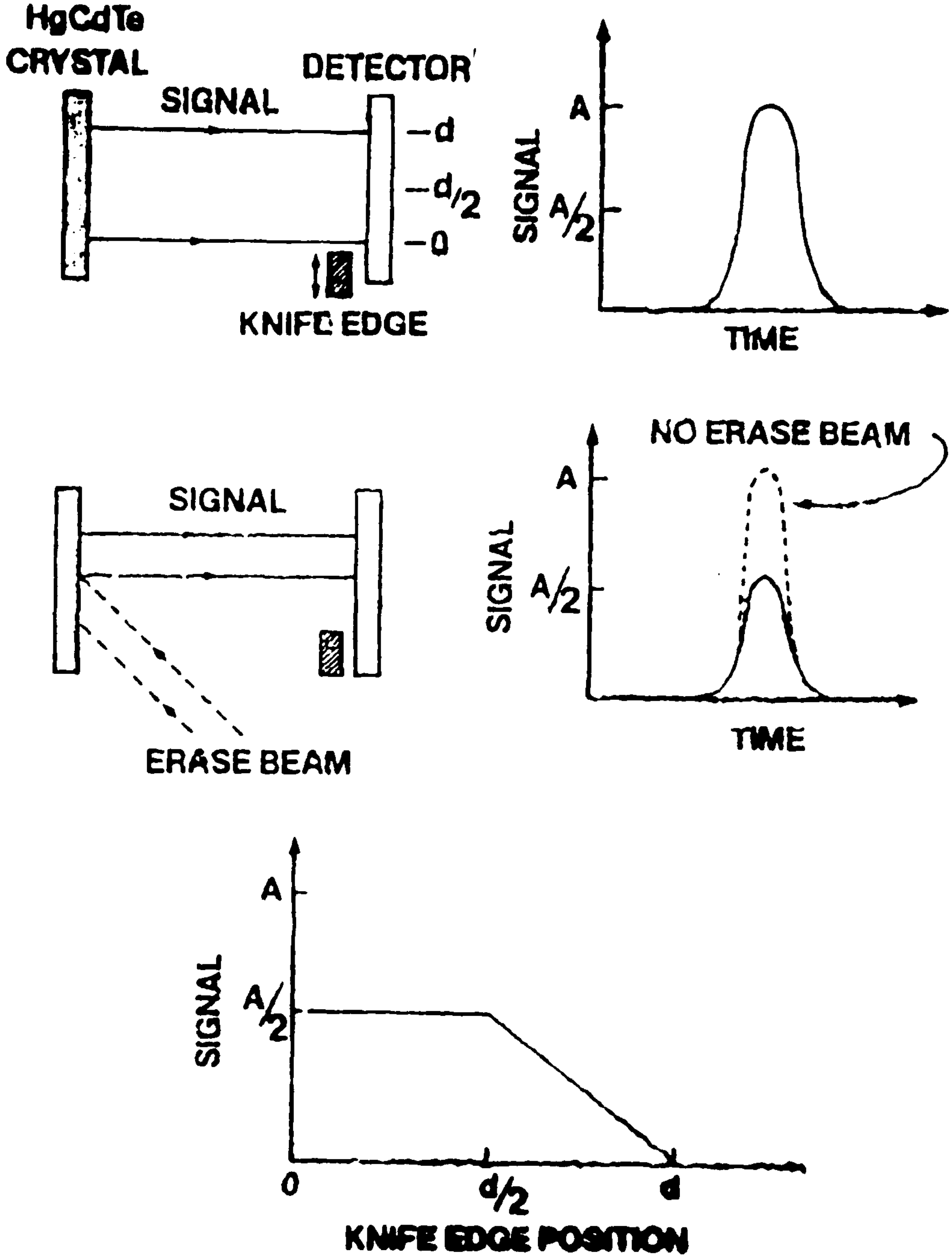


FIGURE 5

Dependence of Diffracted Signal Upon
Spatial Overlap Between Pump/Probe and Erase Beams

Any model must take into account the wavelengths of the $Hg_{0.77}Cd_{0.23}Te$ absorption edge and that of the TEA laser. At 80°K, the absorption edge of $Hg_{0.77}Cd_{0.23}Te$ is at 8.7 μm. The TEA laser emits at 10.6 μm. Until now, it was believed that the only operable nonlinear mechanism under this condition is conduction band nonparabolicity. However, A. Miller et al.(1) have employed a 10.6 μm CO_2 laser to measure two-photon absorption in $Hg_{0.78}Cd_{0.22}Te$ at room temperature, where the absorption edge is at 7.1 μm. They find a two-photon absorption coefficient K_2 of 14cm/MW. Thus radiation from the CO_2 TEA laser employed in our experiments, although of too low energy to produce hole-electron pairs by single photon excitation, produces them with low efficiency by two-photon excitation.

- Photoexcited plasma is the operable mechanism.

Given the presence of hole-electron pairs produced by two-photon excitation, the operable nonlinear mechanism is the photoexcited plasma one. The expression for the third order susceptibility $\chi^{(3)}$ is (2,3)

$$\chi^{(3)} = \frac{\eta\alpha n_o c e^2}{8\pi m^* \hbar\omega^3}; \qquad (1)$$

where η is the quantum efficiency for excitation of hole-electron pairs, α is the absorption coefficient, n_o is the refractive index, c is the speed of light, e is the electronic charge m* is the effective optical mass of a hole-electron pair, $\hbar$ is Planck's constant divided by 2π, and ω is the angular frequency of the radiation. Note that $\chi^{(3)}$ depends linearly upon τ, the effective lifetime of the photoexcited hole-electron pairs.

- Auger recombination dominates.

At 77°K in $Hg_{0.77}Cd_{0.23}Te$ it is known that Auger recombination is the operable mechanism in relatively high purity samples. The expression for the Auger lifetime τ, is given by

$$\tau_A = \frac{2n_i^2\tau_{Ai}}{(n_o+p_o+n_e)(n_o+p_e)}. \qquad (2)$$

Here n_i is the intrinsic concentration, n_o and p_o are the thermally excited concentrations of free electrons and holes, n_e and p_e are the excess, photoexcited electron and hole concentrations, and τ_{Ai} is the Auger lifetime in intrinsic material. Assuming that $n_e = p_e$, and that these concentrations are greater than n_o and p_o, Eq. (2) reduces to

$$\tau_A = \frac{2n_i^2\tau_{Ai}}{n_e^2}. \qquad (3)$$

Thus the Auger lifetime is inversely proportional to the square of the photoexcited carrier concentration if the photoexcitation intensity and efficiency are high.

Hill, Parry, and Miller [4,5] have shown that the dependence of τ_A on the excitation intensity must be taken into account when studying nonlinear optical effects in (Hg,Cd)Te. In experimentally evaluating a $Hg_{0.77}Cd_{0.23}Te$ Fabry-Perot etalon they found it necessary to invoke Eqs. (2) and (3) to explain their data.

- The effect of the erase beam is to increase the recombination rate of the photoexcited electrons and holes, thereby reducing $\chi^{(3)}$.

The erase beam produces hole-electron pairs. This increment to the excess pairs produced by the pump and signal beams reduces the lifetime according to Eq. (3), thereby reducing $\chi^{(3)}$ according to Eq. (1). Since the power in the diffracted beam [6] depends upon $|\chi^{(3)}|^2$, the signal decreases in the presence of the erase beam.

The above tentative explanation needs to be examined in detail before it can be said to be the operable one.

1. A. Miller et al., J. Phys. C. Solid State Phys. 12, 4839(1979).

2. R.K. Jain and M.B. Klein, Appl. Phys. Lett. 35, 494(1979).

3. R.K. Jain and D.G. Steel, Appl. Phys. Lett. 34, 1(1980).

4. J.R. Hill, G. Parry, and A. Miller, Optics Comm. 43, 151(1982).

5. G. Parry, A. Miller, and R. Daley, Rochester Mtg. on Optics, 1983.

6. P.W. Kruse, M.A. Khan, and J.F. Ready, "Optical Phase Conjugation in (Hg,Cd)Te," SPIE 293, Wavefront Distortions in Power Optics, SPIE, Bellingham, Wash., 1981.

2.2 Nonlinear Optics Experimental Investigations - Plans

During the final period of the contract, further experimental investigations will be made of the dynamics of real-time electron gratings. The explanation offered above will be analyzed in greater depth to determine whether it is appropriate. If not, a new mechanism will be sought.

2.3 Nonlinear Optics Theoretical Investigations - Status

Honeywell has entered into an agreement with Prof. Yia-Chung Chang of the Department of Physics, University of Illinois at Urbana-Champaign, to carry out research objectives (9) and (10). This followed from the departure from Honeywell of Dr. Darryl Smith, who was originally to have carried out these tasks.

During the period 9 July 1984 - 8 January 1985 Prof. Chang addressed objective (10), "Investigate theoretically the nonlinear optical interaction mechanisms in semiconductor superlattices." His analysis is contained in the manuscript "Non-Linear Optical Properties of Semiconductor Superlattices," which he has submitted for publication to the Journal of Applied Physics. His findings are summarized below.

Prof. Chang studied the third order susceptibility $\chi^{(3)}$ due to both the conduction band nonparabolicity and the photo-excited plasma mechanisms in both $GaAs/Al_xGa_{1-x}As$ and $Hg_{1-x}Cd_xTe/Hg_yCd_{1-y}Te$ superlattices. His calculation is based upon a two-band k.p model, which he has previously shown gives a fairly good description of energies and wavefunctions of the conduction band states in both GaAs/AlGaAs and HgTe/CdTe superlattices. His analysis shows that with the appropriate choice of superlattice parameters, the optimized $GaAs/A\ _xGa_{1-x}As$ superlattice has a value of $\chi_{(3)}$ due to conduction band nonparabolicity which is about two orders of magnitude larger than the corresponding GaAs bulk value, see Figure 6. However, for a small bandgap $Hg_{1-x}Cd_xTe/Hg_yCd_{1-y}Te$ superlattice the third order susceptibility $\chi^{(3)}$ due to conduction band nonparabolicity is approximately the same as that of bulk $Hg_{1-x}Cd_xTe$, see Figure 7.

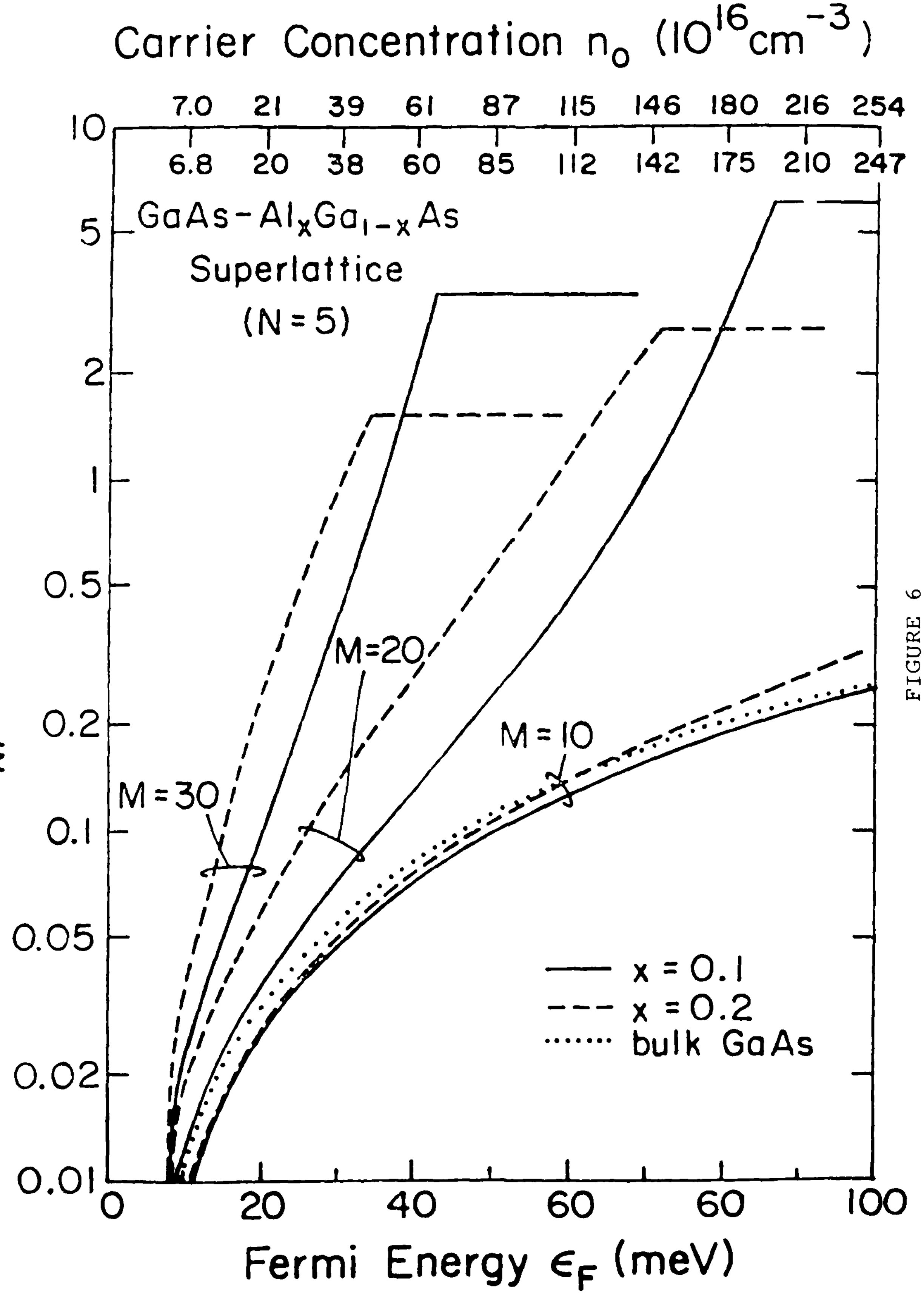


FIGURE 6

Third-order susceptibility due to conduction band nonparabolicity $X^{(3)}_{NP}$ of GaAs-$A\ell_xGa_{1-x}As$ superlattices plotted against the Fermi energy ϵ_F for N = 5, M = 10, 20, 30, and x=0.1, 0.2, and for bulk GaAs. The upper values of n_o are for M=10 and x=0.2. The lower values of n_o are for M=10 and x=0.1. Here M is the number of GaAs atomic layers and N is the number of $A\ell_xGa_{1-x}As$ layers.

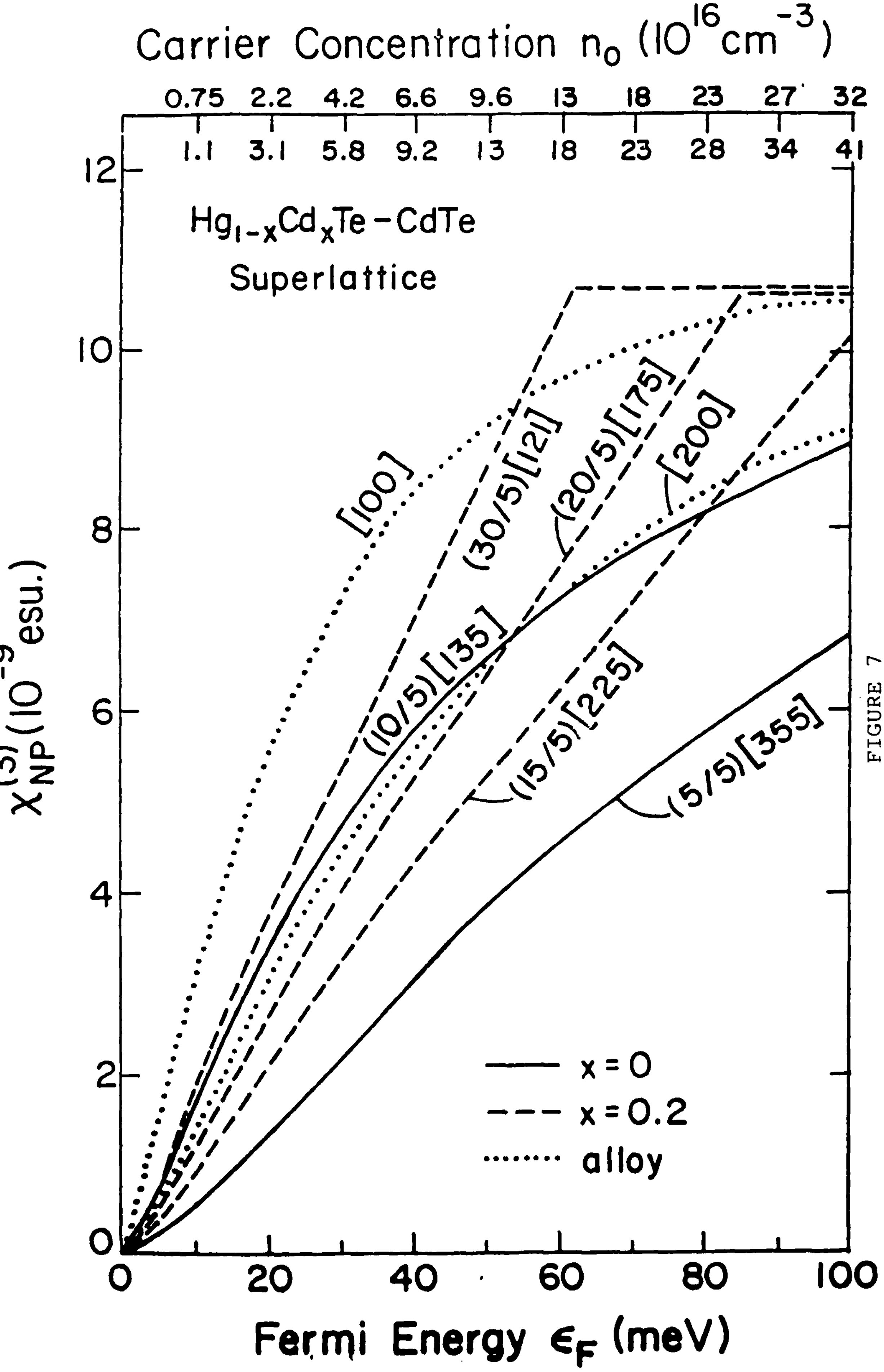


FIGURE 7

$\chi^{(3)}_{NP}$ of a number of $Hg_{1-x}Cd_xTe$-CdTe superlattices labelled by (M/N) and HgCdTe alloys plotted against ϵ_F. The numbers in square brackets denote the fundamental gap energy (in meV). The upper values of n_o are for (M/N) = (30/5) and x=0.2. The lower values of n_o are for (M/N) = (10/5) and x=0. Here M is the number of $Hg_{1-x}Cd_xTe$ atomic layers and N is the number of CdTe atomic layers.

Prof. Chang employed both the direct saturation model and the Burstein-Moss model to determine the influence of the photo-excited plasma mechanism on $\chi^{(3)}$. He found that in both models $\chi^{(3)}$ is related to the squared optical matrix element, the interband relaxation time, and the linear absorption coefficient. These parameters in $GaAs/AlGa_{1-x}As$ and $Hg_{1-x}Cd_xTe/Hg_yCd_{1-y}Te$ superlattices do not deviate substantially from the bulk materials. Thus the value of $\chi^{(3)}$ in a $GaAs/Al_xGa_{1-x}As$ superlattice arising from the photoexcited plasma mechanism is substantially the same in the bulk $Al_xGa_{1-x}As$ alloy. Under the most favorable conditions the value of $\chi^{(3)}$ due to the photoexcited plasma mechanism is about three times higher in the $Hg_{1-x}Cd_xTe/Hg_yCd_{1-y}Te$ superlattice than that of the bulk $Hg_{1-x}Cd_xTe$ alloy.

These results of Prof. Chang have far reaching significance. They show that regardless of mechanism, $Hg_{1-x}Cd_xTe/Hg_yCd_{1-y}Te$ superlattices having small energy gaps are not substantially better in terms of their third order susceptibility than bullk alloys of small gap $Hg_{1-x}Cd_xTe$. Thus those nonlinear optical effects depending on the third order susceptibility which have been investigated for bulk $Hg_{1-x}Cd_xTe$ under this contract, including the spin-flip Raman laser, spin-resonant four-wave mixing, degenerate four-wave mixing, optical phase conjugation, and optical bistability would work no better in $Hg_{1-x}Cd_xTe/Hg_yCd_{1-y}Te$ superlattices than in the bulk $Hg_{1-x}Cd_xTe$ alloy.

On the other hand, Prof. Chang showed that the third order susceptibility arising from conduction band nonparabolicity is substantially higher in $GaAs/Al_xGa_{1-x}As$ superlattices compared with that in the bulk $Al_xGa_{1-x}As$ alloy. Unfortunately there is no improvement in the third order susceptibility due to the photoexcited plasma mechanism in the $GaAs/Al_xGa_{1-x}As$ superlattice compared with the bulk $Al_xGa_{1-x}As$ alloy. Thus experiments which exploit the superior nonlinear optical properties of $GaAs/Al_xGa_{1-x}As$ superlattices should be based upon conduction band nonparabolicity rather than the photoexcited plasma mechanism.

2.4 Nonlinear Optics Theoretical Investigations - Plans

During the next, and last, period of the contract, Prof. Chang will address objective (9), "Investigate theoretically the response time of nonlinear optical interactions produced by the various microscopic mechanisms in semiconductors."

3.0 WRITTEN PUBLICATIONS IN TECHNICAL JOURNALS

One written paper was submitted for publication during this reporting period.

Y.C. Chang, "Nonlinear Optical Properties of Semiconductor Superlattices," submitted to the Journal of Applied Physics.

4.0 PROFESSIONAL PERSONNEL ASSOCIATED WITH RESEARCH EFFORT

The following personnel with B.S. or higher degrees participated in the research effort during this reporting period.

Dr. Paul W. Kruse, Chief Research Fellow
Dr. Muhammad A. Khan, Senior Principal Research Scientist
Dr.David K. Arch, Principal Research Scientist
Mr. John A. Lehman, Student Aide
Prof. Y.C. Chang, Assistant Professor, University of Illinois.

5.0 INTERACTIONS

5.1 Spoken Papers Presented at Meetings

One spoken paper was presented during this reporting period.

M. Asif Khan, J. Lehman, and P.W. Kruse, "Dynamics of Real-Time Electron Gratings in $Hg_{1-x}Cd_xTe$," presented at International Conference on Lasers '84, San Francisco, 26-30 November 1984.

5.2 Consultative and Advisory Functions

None

5.3 Other Interactions

None

6.0 NEW DISCOVERIES, INVENTIONS, OR PATENT DISCLOSURES

None